

In vitro, Appraisal and abatement of drainage wastewater pollution in light of utilizing fly ash

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Abstract: An examination was conducted to assess and provides spatial variation in drainage water quality parameters along Janag drain, Kafr El-Zayat area, El-Gharbia Governorate, Egypt, with respect to Law 48 of 1982 and its modification in 2013. Abatement of inorganic and organic pollutants using fly ash as well as to investigate the removal efficiency of fly ash based adsorption. Collected and treated water samples were analyzed to determine chemical and microbiological parameters. Fly ash filters were prepared by two approaches and the effects of flow rate on removal efficiency have been studied. The results indicated that the mixed drainage samples showed significant variation for all water quality parameters studied except pH with respect to drainage, industrial and sewage wastewater. BOD, COD, TN, TP and total Coliforms are higher than the permissible limits for drainage water. The results of treatment by fly ash briquette bed proved that fly ash was capable of reducing BOD, COD, TN and TP by 77%, 90%, 59% and 100 %, respectively at flow rate 0.5 l/min to lower than the recommended limits. The chlorine dose required for purifying water treated with fly ash approaches was 9 to 12 mg/l, respectively. Further studies required in situ for treating wastewater before discharge into fresh water bodies using fly ash briquette bed approach.

[Lubna A. Ibrahim, H.A.A El Gammal and Belal N.A. Mahran. **In vitro, Appraisal and abatement of drainage wastewater pollution in light of utilizing fly ash.** *Nat Sci* 2017;15(12):79-89]. ISSN 1545-0740 (print); ISSN 2375-7167 (online). <http://www.sciencepub.net/nature>. 7. doi: [10.7537/marsnsj151217.07](https://doi.org/10.7537/marsnsj151217.07).

Key words: Fly ash; drainage; industrial; domestic; water quality; Adsorption.

1. Introduction

Reusing wastewater affords Egypt of the solutions to the detrimental impact of water shortage on its agricultural extension. The degraded quality of wastewater creates a serious environmental danger which at last constraints its reuse. Several legislative and institutional measures have been taken to improve the wastewater quality and keep the environment (Howsam and Carter, 1996). These measures have not resulted in clean water, since they concentrated on technical prerequisites and forcing sanctions on offenders rather than finding economics uses for the produced pollutants (Khouzam, 1996). In the present study uses fly ash for treatment of wastewater and find a benefit to fly ash powder.

In Egypt, the greater part of the villages and cities dump their wastes, including human waste in the nearby waterways. No one thought that part of the waste could be returned back through drinking water, washing or crops grown with this water. Water pollution not only effects on public health, but also on the economic factors important to water quality and natural resources of reusable waters (Abdel-Shafy and Aly, 2002). This situation occurred in the areas where Janag drain passes through Kafr El-Zayat area, El-Gharbia Governorate, Egypt. The residents threw all their wastes, including household and sanitation, dumping of domestic wastewater from sewage station, in addition to industrial wastewater from Alexandria

Oil and Soap (factory in Kafr El-Zayat). That results in the gathering of large amounts of pollutants in the drain and its aspects. The most pronounced pollutants in the oil processing wastewaters are suspended solids, pH, organics, sulfide, muds, brine, and ammonia. The excessive discharge of wastewater to sewerage systems brought about the clogging of sewers and pumping plants and obstruction of biological treatment processes (Abou-Elela and Zaher, 1998). So, it is imperative to find an approach to solve the problem of drainage wastewater quality. In the present investigation Janag drain wastewater was selected and two approaches for treatment were applied on laboratory scale.

There are many chemical methods have been employed for the elimination of heavy metals, COD and color from industrial effluents and drainage wastewater (Robinson *et al.*, 2001). Chemical coagulation (Ewida and Ibrahim, 2014), fenton reaction (Hsueh *et al.*, 2005), precipitation (Linstedt *et al.*, 1981), sorptive flotation (Ghazy *et al.*, 2001), ultra-filtration, nano-filtration (Mo *et al.*, 2008), ion exchange (Jester, 1973), reverse osmosis (RO), adsorption (El-Sharkawy *et al.*, 2001); Aklil *et al.* (2004); Youssef *et al.* (2004); Jang *et al.* (2005); Zhou, *et al.* (2004); Kadirvelu *et al.* (2001)), and electrochemical techniques (Chen, 2004) are among the variety of approaches which are utilized for the treatment of wastewater. Adsorption is found

successful for the removal of pollutants from wastewater. It is simple, cost effective, quick and very effective process if a low cost adsorbent is utilized (Kamboh *et al.*, 2009).

Fly ash is produced in an immense amount as a waste from the utilization of coal in power generation. Fly ash was mainly composed of SiO_2 , Al_2O_3 & Fe_2O_3 as major components and CaO , K_2O , Na_2O , SO_3 & MgO as minor components (Ibrahim, 2015). Using fly ash to treat wastewater is an environmental achievement contributes to the utilization of waste, and is an important economic step because it lessens the costs of wastewater treatment. Several previous studies have demonstrated (Ahmaruzzaman 2009; 2010; Shah *et al.* 2015; Sanas *et al.*, 2016; Adegoke *et al.*, 2017) that fly ash could mitigate or reduce various pollutants from wastewater.

The water characteristics of Janag drain necessitate amelioration to meet the requirement of Egyptian guidelines (Ministerial Decree No. 92 of 2013 amending the Ministerial Decree No. 8 of 1982 on the executive Regulations of Law No. 48 of 1982 concerning the Protection of the Nile River and water channels from pollution). The present investigation is taken in light of the fact that there is no work done on Janag drain and just a little work for using of fly ash as adsorbent material for treating drainage wastewater. The present examination was conducted to study the quality of drainage, industrial and sewage wastewater collected along Janag drain. Study spatial variation between examining locations along Janag drain in order to determine the impact of these wastes on the drain. Preparation of two approaches on laboratory scale; the first approach consisted from fly ash powder fixed by fabric material in a plastic bottle, while the second approach consisted from Jar test coupled by prepared fly ash briquette bed. Determine the amount of chlorine required for disinfection. A comparison between these approaches for treatment wastewater from a Janag drain in order to meet the prerequisites under the environmental legislations of Egypt was done.

2 Materials and Methods

2.1. Study area

Janag drain is one of the drains, which passes between Basion and Kafr El-Zayat, El-Gharbia Governorate, Egypt. The length of Janag drain is about 17.4 km and serves 5000 acres in addition to flow from other branch drains. Janag drain discharges into Tahwelet Nemra 9 Ela'la drain. The following drains release in Janag drain: Qunah drain (length of the drain is 17 kilometers, Serves the reins of 15600 acres), Mahalet Diay drain (length of the drain is 3 kilometers and 900 meters, Serves the reins of 2300 acres) and Meneit Janag drain (length of the drain is 3

kilometers and 400 meters, Serves the reins of 2800 acres). Janag drain receives high concentrations of organic compounds and nutrients. The major sources of pollution on the drain are industrial wastewater from Alexandria Oil & Soap factory in Kafr El Zayat Figure 1A, domestic wastewater from the sewage station of Kafr El Zayat, Figure 1B and encroachment with household waste and sewage, Figure 1C.

2.2. Wastewater sampling

A sum of 24 water samples was collected from Janag drain and outlets of oil & soap factory and sewage station on Janag drain, in the period from August 2016 to April 2017. The samples collected from six different locations as described in table 1 & figure 2. To give spatial variation in drainage water quality along Janag drain prior and after blending with industrial and domestic wastewater. Water samples were collected in polyethylene containers for physico-chemical analysis and sterilized glass containers for bacteriological analysis. The samples were filtered by the filtration system (through a membrane filter of pore size 0.45μ) and acidified with nitric acid to $\text{pH} < 2$, to prevent any change in the chemical composition of the sample before analysis of major cations and trace elements. Sterilization was made by sodium thiosulfate 0.1 ml of 3% $\text{Na}_2\text{S}_2\text{O}_3$ solution in 120 ml bottle. For treatment methods, 6 bottles were collected, each of them have 20 litre wastewater sample. They were immediately transported in a water cooler at 4°C to Central Laboratory for Environmental Water Quality Monitoring (CLEQM).

2.3. Reagents and Quality assurance

Analytical chemicals grade and deionized water were used for preparing all solutions. All plastic bottles were cleaned by soaking in 10% HNO_3 solution, rinsing with deionized water, and drying in an oven. Procedural blanks, preparation of standard solutions under clean laboratory environment. Triethanolamine grade, 98% ($\text{C}_6\text{H}_{15}\text{NO}_3$) was an Oxford Laboratory product, Mumbai, India. A sodium hypochlorite solution was a Loba Chemie product, India. Stock standard solutions of cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn) and zinc (Zn), were obtained from Merck in concentrations of 1000 mg/L (Merck, Darmstadt, Germany). Recovery studies for the trace and major elements analysed using ICP-OES & ICP-MS (inductively coupled plasma-optical emission spectrometry & inductively coupled plasma-mass spectrometry) ranged between 95 and 105%. All calibration standards were prepared from 1000 mg/l stock solutions of each metal using further dilution. All the experiments were repeated thrice, and the mean value & standard deviation (SD) obtained are being reported.

2.4. Water quality determination

The parameters (pH, TDS, TSS, TN (total nitrogen), TP (total phosphorus), BOD, COD, Cd, Cr, Cu, Fe, Mn, Pb, Zn and TC (total Coliforms)) were selected relying on decree 92 (2013) of Egyptian Law

48. While, cations and anions were measured in spite of the fact that they don't consider in the Law to decide the reason of deteriorating or improving TDS. To determine the forms of TN in samples nitrite, nitrate and ammonia were measured.



Figure 1. Sources of pollution along Janag drain, while A: Alexandria oil & soap factory in Kafr El Zayat, B: Sewage station of Kafr El Zayat and C; household waste in Janag drain.

Table 1. Sampling sites description.

Code	Description
DR1	Drainage water samples collected from Janag drain (1 Km before oil & soap factory)
IS2	Industrial wastewater collected from the outlet of pipe 1 (oil & soap factory).
IS3	Industrial wastewater collected from the outlet of pipe 2 (oil & soap factory).
SW4	Municipal water collected from the outlet of sewage station of Kafr El Zayat.
DR5	Drainage water samples collected from Janag drain (1 Km after sewage station).
DR6	Drainage water samples collected from Janag drain (5 Km after sewage station). In the treatment process , water collected from this site.

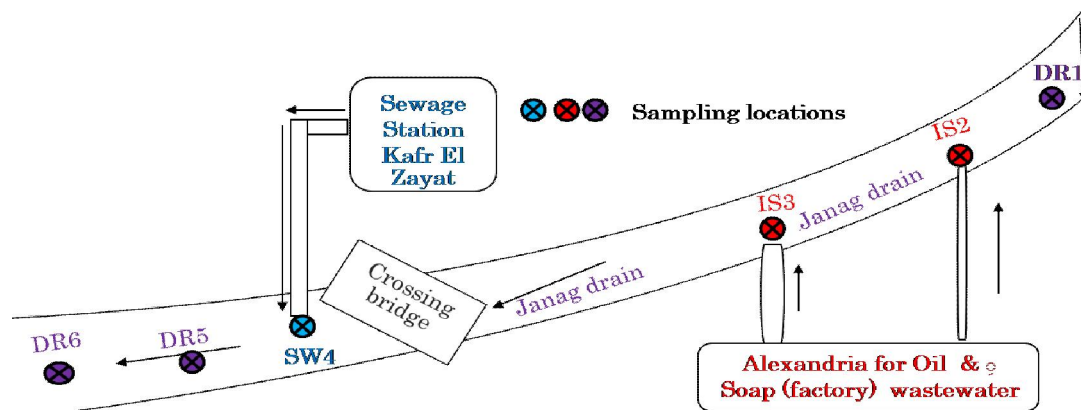


Figure 2. The modeling area, 3 (DR1, DR5 and DR6) drainage, 1 (SW4) sewage and 2 (IS2 & IS3) industrial wastewater sampling locations along Janag drain.

Chemical analyses: water characteristics were determined according to standard methods for testing fresh water and wastewater (APHA *et al.*, 2012). pH was measured at 25°C using Info Lab meter. Carbonate (CO₃²⁻) and bicarbonates (HCO₃⁻) ions were determined titrimetrically against 0.2 N-H₂SO₄, using phenolphthalein and methyl orange indicators, respectively. Ammonia & TN were determined by

Kjeldahl Method. TP was determined colorimetric detection using continuous flow analysis after digestion with alkaline persulphate (Patton and Kryskalla, 2003). Total dissolved solids (TDS) were determined by weighing the solid residue obtained by evaporating a measured volume of filtered water sample to dryness at 103-105°C. Suspended solids were determined by filtering a known volume of water

through a 0.45 µm filter paper and noting the increase in weight of the filter paper after dryness at 103-105°C. Major anions; chloride (Cl⁻), sulfate (SO₄²⁻) and nitrate (NO₃⁻) were measured using Ion Chromatography (IC). Major cations; calcium (Ca²⁺), potassium (K⁺), magnesium (Mg²⁺) and sodium (Na⁺) were measured by inductively coupled plasma-optical emission spectrometry (ICP-OES). Heavy or trace elements (Cd, Cr, Cu, Fe, Mn, Pb and Zn) were measured by inductively coupled plasma-optical Emission spectrometry (ICP-OES) Dual View.

Biological Oxygen Demand (BOD) was measured using BOD fast respirometry system model TS606/2 at 20°C incubation in a thermostatic incubator chamber model WTW for 5 days (APHA *et al.*, 2012). The Chemical Oxygen Demand (COD) was measured calorimetrically using a strong chemical oxidant (potassium dichromate) in acid medium. The mixture was heated to oxidize organic carbon to carbon dioxide and water. It measures the amount of dichromate consumed in the breakdown of organic matter using COD Reactor (block heater operates at 150±2°C) and Spectrophotometer model Huch-DR-2010 (APHA *et al.*, 2012).

Microbiological Analyses: Collected and treated wastewater samples were examined within 6 hours, according to standard methods (APHA *et al.*, 2012) for enumeration of total Coliforms (TC). Membrane filter technique was applied using a filtration system completed with stainless steel autoclavable manifold and oil-free vacuum/pressure pump for counting fecal Coliforms. All samples were filtered through sterile, surface girded membrane of pore size of 0.45 µm with a diameter of 47 mm. The data were recorded as Colony Forming Unit (CFU/100 ml) using the following formula:

$$\frac{\text{Colonies}}{100\text{mL}} = \frac{\text{Counted colonies}}{\text{mL of sample filtered}} \times 100$$

2.5. Fly ash and treatment methods

Collection and characteristics: Fly ash was collected from Construction Research Institute (CRI), National Water Research Center (NWRC), Egypt. Pipette method (Miller and Miller, 1987) was used to determine the texture of fly ash. pH and EC were measured in the extracted water with a ratio 1 fly ash: 5 water after equilibration for 30 min. Organic matter was determined by an oxidation method (Alexiadis, 1972).

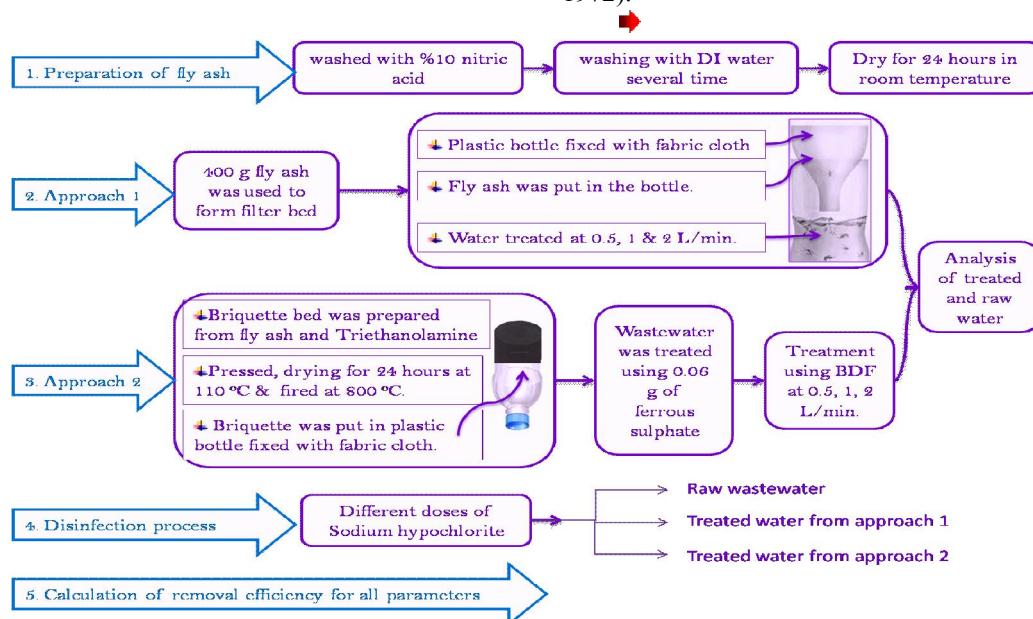


Figure 3. Schematic diagram showing the approaches adopted for preparation and investigating the ability of fly ash for wastewater treatment.

Treatment methods: This section contained five stages, which are, preparation of fly ash, approach 1, approach 2, disinfection process and removal efficiency calculation. The stages were delineated in figure 3 and each one was interpreted as follows:

➔ Preparation of fly ash: Fly ash was cleaned by 10% of HNO₃ for 12 hours, after that washed several times with deionized water to eliminate the

surplus amount of acid. Fly ash was dried at room temperature for 24 hours. The aim of acid treated fly ash was to remove of organic material, available elements and improve the adsorption capacity of fly ash. Since the adsorption capacity for acid treated fly ash was observed to be 2.4x10⁻⁵ mol/g, while non-treated fly ash demonstrated an adsorption capacity of 1.4x10⁻⁵ mol/g (Wang *et al.*, 2004). The

dried fly ash was used for preparation of filters in the following approach 1 & 2.

➤ Approach 1 (Filter Fly ash powder (FF)): 400 g of fly ash was fixed by fabric cloth in bottle plastic water. Four litres of wastewater samples were passed through the filter at various rates 0.5, 1 and 2 l/min. Then, the treated water (FF) and raw wastewater samples were analyzed for all parameters in the water quality determination.

Approach 2 (Jar test followed by filtration using Briquette bed (CBD)):

❖ Jar test procedure: wastewater was treated by means of coagulation utilizing ferrous sulfate as coagulant. Different doses (0.01, 0.03, 0.06, 0.09 g of ferrous sulfate) were applied to 1 liter of wastewater. The maximum speed was kept up to 145 rpm, mixing period 5 minutes and settling time 60 minutes. After settling, the samples were subject for complete analyses to determine the optimal dose (i.e. at this dose higher removal efficiency happened). The optimum dose was = 0.06 mg/l.

❖ Preparation of Briquette bed (BD): 100 g of fly ash was used to prepare briquette. The briquette consists from fly ash and binding material (Triethanolamine (TEA)) with a ratio 10 g fly ash to 1 ml TEA. The binding material was selected depending on the U.S. patent published by McKee (1953). Fly ash was mixed with TEA to form a homogeneous paste, which was pressed by 2 tons for 1 minute (McKee, 1953) using a manual uniaxial hydraulic press and a steel cylindrical mold. Prepared briquette of fly ash was dried in an oven for 24 hours at the 110°C (Shah *et al.*, 2015) to increase its adsorption capacity. Then, it was fired in Muffle furnace at 800°C for carbonization (McKee, 1953), after that left to cool in desiccators (Shah *et al.*, 2015).

❖ Briquette bed (BD) was fixed by fabric cloth in a plastic bottle for filtration process.

❖ Treated water from Jar test was passed through briquette bed (BD) at different flow rates 0.5, 1 and 2 l/min.

❖ The treated water (CBD) and blank raw water were analyzed for all parameters in the water quality determination.

➤ Disinfection process: The two samples collected after treatment by fly ash in the two approaches were treated with different doses of sodium hypochlorite. Chlorination experiments were carried out using sodium hypochlorite (NaClO). 500 ml of samples were distributed to 500 ml sterile beakers. Sodium hypochlorite was spiked into the sample to establish desired doses of chlorine of 5, 10, 15, 20, 25, and 30 mg/l, respectively. The concentrations of chlorine were analyzed by the DPD method (Environmental Protection Agency of China, 2002). After chlorine was added, the samples were

stirred immediately to mix and allowed a contact time of 30, 60 and 90 min to investigate the effect of time on disinfection. The sodium thiosulfate solution (Na₂S₂O₃, 1.5 %, (w/v)) was added into the samples to terminate chlorination. After treatment the sample subjected to analyses to determine total Coliforms.

➤ Removal efficiency: The removal efficiency was determined according to the following formula, where *C_i* and *C_f* are the initial and final concentrations of waste and treated waters, respectively.

$$\text{Removal efficiency (\%)} = \left[\frac{C_i - C_f}{C_i} \right] \times 100$$

2.6. Programs used for interpretation and reporting

Basic descriptive parameters (mean values and standard deviation (SD)) were obtained using SPSS, ver. 15, 2006, statistical software. Analysis of variance (ANOVA) and multiple component tests were applied to all parameters to identify significant differences (*p*<0.05), using Costat statistical program, ver. 6.400, USA.

3 Results and Discussion

Samples collected from six locations were analyzed in triplicates and the relative standard deviation was under 5%. The statistical analyses of chemical and microbiological parameters of samples were summarized in table 2. The results of the treatment process were organized in table 3 and depicted in figures 4 & 5.

3.1. Wastewater quality

The obtained results were compared to the permissible limits of the modification of the Law of Ministry of Irrigation and Water Resources (48/1982) by decision Number 92, 2013 as shown in table (2). Samples DR1, DR5 and DR6 from the Janag drain were compared with article 51 regarding the limits in the drainage water before mixing with the freshwater bodies, while sample SW4 was compared with article 52 concerning licensed criteria throwing municipal water to non fresh surface water. Samples IS2 and IS3 were compared article 52; states licensing criteria throwing treated industrial liquid waste to non fresh surfaces. The high value of standard deviation for each parameter indicated results fluctuation which could be attributed to different types of the source of the discharged water and sort of treatment at the sewage station as well as processed oils, operating conditions and washing water during & after the production.

The pH of Janag drain demonstrated non significant variation (*p*>0.05) after exposure to industrial and municipal wastewater. On the other hand, the pH of industrial (IS2 & IS3) and sewage (SW4) wastewater showed significant variation (*p*<0.05) with respect to drainage water (DR1, DR5 & DR6). pH values of all samples were in the

recommended limits of articles 51 & 52, except sample IS2 was less than the recommended limit. Two sorts of wastewater (IS1 & IS2) are generated from oil & soap factory; acid and alkaline. The acid wastewater (IS1) is created from the soap-stock splitting process, which may be attributed to the utilization of sulfuric acid for treating soap during the separation of fatty acid, which produces highly acidic oily wastewater (Decloux *et al.*, 2007). Whereas the alkaline wastewater (IS3) is generated from all the factory's process installations and equipment (Chatoui *et al.*, 2016) could be ascribed to usage of sodium hydroxide for treating fatty acid producing alkaline wastewater containing fatty acids, sodium chloride and heavy or trace metals. The range of pH for acid and alkaline industrial wastewater are compatible with by Eroglu *et al.* (2008) and Chatoui *et al.* (2016), respectively.

Alkalinity in water results from bicarbonate and carbonate ions. Carbonate (CO_3^{2-}) ions were detected at concentrations lower than the detection limit (<0.2) in all samples except sample IS3. The presence of carbonate in sample IS3 could be attributed to the industrial wastewater containing soda from treating soap. In other words, the increase of pH beyond 8.3, the bicarbonate ion is converted to carbonate ion. Bicarbonate ions increased with significant spatial variation ($p<0.05$). Increasing alkalinity could be attributed to bacterial growth on BOD releases CO_2 , which subsequently dissolves in water to yield carbonic acid (H_2CO_3). This rapidly dissociates to bicarbonate ion, increasing the alkalinity (Fekhaoui and Pattee, 1993).

The increasing of TDS in Janag wastewater was significantly different ($p<0.05$) after mixing with industrial and sewage wastewater. Sample IS2 was higher than the recommended limit in article 52, that could be ascribed to salts of sodium sulfate and sodium chloride utilized in soap separation. Sample DR5 was higher than the limit in article 51, while sample DR6 was under the limit. This implies that the deterioration of TDS was diminished after 5 Km from introducing sewage and industrial wastewater sources. The increasing of TDS resulted from the increase of calcium, sodium, magnesium, potassium, chloride, sulfate and TP concentrations significantly ($p<0.05$) in wastewater after blending with industrial and sewage wastewater. The high content of sulfate in IS2 sample ascribed to the use of sulfuric acid during degradation of soap stack in the neutralization process (Verla *et al.*, 2014). The increase of TP in sample IS2 attributed to the utilization of phosphoric acid for removal of phospholipids and lipoproteins in the degumming step (Chatoui *et al.*, 2016).

TSS exhibited a significant spatial variation ($p<0.05$) demonstrating that the increase of TSS after blending with industrial wastewater was significantly

varied ($P<0.05$). High concentrations of suspended solids increase turbidity, in this way restricting light entrance (hindering photosynthetic activity). Suspended solids were results from the presence of particles in suspension, organic materials. The outcomes obtained for industrial wastewater in this study are in the scope of values obtained by Bouknana *et al.* (2014).

The results of BOD and COD indicated that wastewater before and after blending with industrial and sewage wastewater contained a high load of organic matter. Both of BOD and COD data demonstrated significant spatial variation ($p<0.05$). Samples DR1, DR5 and DR6 were higher than the limit of article 51, while samples IS2 and IS3 were higher than the limit of article 52 and sample SW4 was at the limit of article 52. The higher of BOD and COD could be ascribed to heavy organic and inorganic loading. Comparison of COD/BOD ratios of this study with the rules displayed in the studies of Bouknana *et al.* (2004) and Chatoui *et al.* (2016), the outcomes exhibit that;

- COD/BOD ratios for DR1 & SW4 samples were 1.4 & 1.8, which indicate that these wastes are readily biodegradable.
- COD/BOD ratios for DR6 sample was 3, which are considered relatively high and indicates the presence of persistence and/or toxic compound. Persistent untreated, may contaminate receiving water bodies, and increase the environmental risks and cost of sludge treatment and disposal.
- COD/BOD ratios of IS2, IS3 & DR5 samples were found to be 13.1, 5 & 4.7, which are characterized by a high rate of oxidizable substances, fats, fatty acids, sulfates and phosphates as well as low pH values, would be less biodegradable (cause destruction of micro-organisms). The same result for industrial waste water was obtained by Chatoui *et al.* (2016).

These illustrated that industrial wastewater changed the chemistry of Janag drain from biodegradable to relatively high and hardly biodegradable.

Three forms of nitrogen are commonly measured in water bodies which are ammonia, nitrates and nitrites. Total nitrogen (TN) is the sum of total Kjeldahl nitrogen [ammonia, organic (i.e. amino acids, humic acids, proteins & urea) and reduced nitrogen] and nitrate-nitrite. TN demonstrated significant spatial difference ($P<0.05$) after blending with wastes. The concentrations of nitrite and nitrate in drainage, domestic and industrial wastewater were less than detection limit (DL $<0.2\text{mg/L}$) of the instrument, Table (2). This implies that the forms of total nitrogen are ammonia and organic in the studied samples. The low concentration of nitrite and nitrate could be

attributed to the consumption of oxygen for decomposing organic matter and the oxidation of chemical constituents (Boyd, 1990). The higher concentration of ammonia in wastewater could be ascribed to the microbiological decomposition of nitrogenous compounds in organic matter and decay of discharged organic waste released specifically into

water bodies by some industrial processes (IS2, IS3) or as a component of domestic (SW4) sewage (Ibrahim and Ramzy, 2013). The highest concentration of TN in industrial wastewater was clarified by Tidjanhisseine *et al.* (2016) that the soap and oil factories use a high quantity of organic matters to produce soap and vegetable oil.

Table 2: Spatial varieties of studied water quality parameters (mean±SD) along Janag drain as well as compared with the articles 51 & 52 of Egyptian Law 48.

	DR1	IS2	IS3	SW4	DR5	DR6	Article 51	Article 52 municipal	Article 52 industrial
	Mean ±SD						Decree 92 of Law of 48 in 2013		
pH	6.53 ^{±0.2}	1.63 ^{±0.1}	8.87 ^{±0.1}	7.29 ^{±0.2}	6.8 ^{±0.2}	6.79 ^{±0.1}	6.5-8.5	6-9	6-9
Carbonate	<0.2 ^B	<0.2 ^B	43 ^{A±2.3}	<0.2 ^B	<0.2 ^B	<0.2 ^B	-	-	-
Bicarbonate	201.8 ^{±11.3}	<0.2 ^B	188.8 ^{±9}	298.5 ^{±9.5}	242.8 ^{C±15.5}	491.7 ^{A±8.5}	-	-	-
TDS	629.3 ^{±55.2}	14081.8 ^{A±772}	592.3 ^{±18}	860.5 ^{±31.2}	1079.3 ^{B±54.8}	841 ^{±5}	1000	2000	2000
TSS	55 ^{D±4.6}	186 ^{A±12.3}	52 ^{D±3.6}	30 ^{B±4.4}	80 ^{B±6.8}	61 ^{C±5.5}	-	50	50
BOD	175 ^{B±20.8}	71 ^{D±2.5}	690 ^{A±76.3}	44 ^{±5.1}	138 ^{C±26.5}	90 ^{D±9.1}	30	60	60
COD	246 ^{±74.4}	929 ^{±49.8}	3463 ^{A±133.7}	80 ^{±6.2}	645 ^{C±46.1}	269 ^{D±9.4}	50	80	80
Ammonia	19.8 ^{±8.3}	28.2 ^{±4.5}	25.5 ^{B±3.6}	50.9 ^{A±4.9}	51.1 ^{A±7.6}	32.5 ^{B±6.4}	-	-	-
TN	32.7 ^{±8.3}	31.8 ^{±4.5}	29.5 ^{B±3.6}	50.9 ^{A±4.9}	51.1 ^{A±7.6}	35.5 ^{B±6.4}	15	-	-
Calcium	50 ^{±1}	94.5 ^{A±1.4}	15.6 ^{±0.8}	57.4 ^{±4.4}	65.3 ^{±1.5}	78.3 ^{B±1.7}	-	-	-
Potassium	17.5 ^{C±1.3}	11.4 ^{D±1.1}	7.5 ^{±0.6}	28.3 ^{±0.6}	24.5 ^{B±0.6}	28.9 ^{A±0.9}	-	-	-
Magnesium	17.2 ^{±1.9}	29.8 ^{±1.2}	26 ^{±0.8}	28.5 ^{±1.7}	27.1 ^{±1.4}	19.4 ^{±0.4}	-	-	-
Sodium	104.5 ^{±2.9}	1150 ^{A±57.2}	148.3 ^{±5.4}	150.5 ^{±5.8}	251.3 ^{B±6.3}	152 ^{C±2.4}	-	-	-
Chloride	93.4 ^{±2.1}	440.5 ^{A±21.6}	141.5 ^{D±6.1}	204.3 ^{±11}	338.2 ^{B±0.2}	110 ^{±4.5}	-	-	-
Nitrate	<0.2 ^B	<0.2 ^B	<0.2 ^B	3 ^{A±1.2}	<0.2 ^B	<0.2 ^B	-	-	-
TP	7.4 ^{±0.9}	25.1 ^{A±1.3}	6.2 ^{±1.1}	9.2 ^{±1.3}	8.5 ^{±1}	16 ^{B±1.4}	3	-	-
Sulfate	114.2 ^{±3.4}	6684.4 ^{A±170.8}	97 ^{±2.6}	63.5 ^{B±1.3}	143.1 ^{B±1.7}	62.3 ^{B±2}	-	-	-
Cadmium	0.002 ^{A±0.001}	0.002 ^{A±0.001}	0.002 ^{A±0.001}	<C	0.001 ^{B±0.001}	<C	0.03	0.003	0.003
Chromium	0.078 ^{A±0.007}	0.024 ^{B±0.002}	0.018 ^{B±0.001}	0.023 ^{B±0.001}	0.02 ^{B±0.002}	0.011 ^{±0.002}	0.05	0.1	0.1
Copper	0.026 ^{D±0.002}	0.076 ^{C±0.003}	0.055 ^{±0.002}	0.11 ^{A±0.011}	0.032 ^{D±0.002}	0.047 ^{±0.007}	1	0.5	0.5
Iron	0.909 ^{B±0.081}	10.81 ^{A±0.573}	0.619 ^{B±0.028}	0.836 ^{B±0.033}	1.044 ^{B±0.059}	0.78 ^{B±0.05}	3	3.5	3.5
Lead	0.079 ^{B±0.007}	0.546 ^{A±0.029}	0.016 ^{±0.001}	0.01 ^{C±0.001}	0.017 ^{±0.001}	<C	0.01	0.1	0.1
Manganese	0.133 ^{±0.012}	0.522 ^{A±0.03}	0.337 ^{±0.013}	0.453 ^{B±0.016}	0.448 ^{B±0.024}	0.396 ^{±0.02}	2	-	-
Zinc	0.097 ^{B±0.008}	0.427 ^{A±0.023}	0.102 ^{B±0.004}	0.014 ^{±0.001}	0.012 ^{C±0.001}	0.026 ^{±0.001}	2	2	2
T. Coliform	875×10 ³ ±273×10 ⁴	6×10 ³ ±1×10 ⁴	32×10 ³ ±8×10 ⁴	68×10 ³ ±14×10 ⁴	640×10 ³ ±57×10 ⁴	700×10 ³ ±82×10 ⁴	5000	5000	5000

* pH is unitless, T. Coliform (TC) unit is CFU/100ml, while the units of other parameters in table 2 are mg/L. Data represented as mean ± SD of 4 samples. Different single letters (A, B, C, D, E, F) in the same row are significantly different ($p < 0.05$). Codes DR1, DR5 and DR6 are drainage wastewater, code SW4 is sewage wastewater and codes IS2 and IS3 are industrial wastewater along Janag drain.

The results of heavy or trace elements were less than the recommended limits of articles 51 & 52 with the exception of Fe and Pb in sample IS2 from oil & soap factory. The higher values of these elements could be ascribed to acidity (pH<2) of IS2 sample. The results are in good agreement with Tidjanhisseine *et al.* (2016), they found that industrial wastewater from the soap and oil factories contained Fe, Cu, Pb, Zn and Cr. The reduction of Cd, Cr, Cu, Mn, Pb and Zn after blending with industrial acidic water can be ascribed to the change pH of water from acidic (IS2) to natural (DR5 & DR6) and subsequent precipitation of these elements takes place in the drain. The results thus obtained are compatible with Balintova *et al.* (2012) experiment that the variability of pH influences the sediment-water partitioning of trace elements (Fe, Cu, Zn & Mn).

Total Coliforms (TC) represented in table (2) showed that the highest count in Janag drain water, followed wastewater from sewage station and oil & Soap Company. For total Coliforms, Janag water reduced with significant variation ($p < 0.05$) after blending with domestic and industrial wastewater. Total Coliforms counts in all samples were higher than the permissible limits of articles 51 & 52. Bacterial

contamination recorded in this study could be attributed mostly to domestic sewage pollution and agriculture runoff (Zaghloul and Elwan, 2011).

3.1. Physical characteristic of fly ash

The result indicated that fly ash was alkaline, with pH 10.62 ± 0.02. Electrical conductivity was 111 ± 0.05 μS/cm and organic matter (%) was 1.27±0.1 %. The result of texture analysis showed that fly ash samples comprise fine sand (26.43%±0.03), clay (15.20%±0.02) and silt (58.37%±0.03) indicated that fly ash was silt loam texture (Soil Survey Staff, 1975).

3.2. Treatment process

The outcomes of the present study indicated that the drainage water contains high concentrations of TP, TN, BOD, COD and TC. Furthermore, investigated the efficacy of fly ash to reduce these pollutants from wastewater. Two approaches using fly ash were carried on wastewater; adsorption treatment followed by a chlorination process was used to treat mixed wastewater.

Comparison between the two approaches for using fly ash for treatment: The obtained results of the treated water were in comparison to the permissible limits of article 51 concerning the license criteria of releasing the drainage water before

throwing to the freshwater bodies as appeared in table 3. The impact of fly ash strategies on the chemical and microbiological water quality parameters of Janag drain is well studied and the results are summarized in table 3, with the following notes;

Statistically, the results of table 4 demonstrated that the two strategies to remedy of drainage water showed significant difference ($P < 0.05$) for all studied water quality parameters except sodium, chloride, cadmium and lead. In order that the effect of the two approaches on pH, alkalinity, TDS, TSS, BOD, COD, ammonia, TN, TP, calcium, potassium, magnesium, sulphate, TC, chromium, copper, iron, manganese and zinc are unmatched.

FF (Filter fly ash powder) reduced BOD, COD, TN, TP and TC with significant variation ($p < 0.05$). The removal efficiency for BOD, COD, TN, TP and total Coliforms was 61.09%, 61.93%, 47.21%, 96% and 85.7%, respectively. The values of BOD and COD are smaller than the values obtained by Sanas *et al.* (2016) which was 71.48% and 66.59% in 10 cm thick fly ash, respectively. The disparity could be ascribed to the nature of waste, wastewater to fly ash ratio as well as the rate of flow.

FF failed to reduce BOD and COD to the prescribed values 30 and 50 mg/l (recommended limits), respectively. Significant elevated variation ($p < 0.05$) for pH, TDS, calcium and magnesium were observed. The extent of pH and TDS were still within the recommended range of article 51 of Law 48, while calcium and magnesium weren't considered in article 51 of Law 48. The increase of TDS could be related to increase of calcium and magnesium leached from the fly ash filter (FF) in the course of treatment as treatment residue.

CBD (Briquette bed coupled with Jar test) improved BOD, COD, TN, TP and TC concentrations with significant difference ($p < 0.05$). Jar test (coagulation) influenced the reduction of BOD, COD, TN, TP and TC by 25%, 27%, 5%, 60% and 80%, respectively. The CBD coupled with Jar test, resulted an excessive removal efficiency for BOD, COD, TN, TP and TC by 78.22%, 90.48%, 41.35%, 98% and 90%, respectively. The value of COD was higher than the values obtained by Shah *et al.* (2015) which was 67%. The variation could be attributed to the flow rate; in the present study was 0.5 l/min, while the previous study was 1 l/min as well as the dose used for coagulation.

CBD deteriorated concentration of pH, TDS, calcium and sodium with significant variation ($p < 0.05$), while sulphate increased with non significant variation. The ranges of pH and TDS were below the values of article 51 of Law 48, while sulphate, calcium and sodium weren't considered in article 51 of Law 48. The increase of sulphate and

TDS in water after applying CBD approach attributed to the use of ferrous sulphate in the coagulation step, that are in good agreement with the result obtained by Nanda and Vyas (2014). CBD failed to reduce TC to value lower than the recommended value 5000 CFU/100 ml.

The results indicated that fly ash adsorbs a wide range of impurities and contaminants, including BOD, COD, TN, TP, TSS, sulphate, heavy & trace elements and TC. Other substances, like sodium and chloride, are not as attracted to the carbon and are not filtered out. Both CBD and FF could capture organic compounds and trace & heavy metals (cadmium, copper, chromium, lead, manganese and zinc) and suspended solids effectively due to high porosity and surface area, which leads to high adsorption capacity. The removal of wastewater contaminant by fly ash is owing to physical process called adsorption. The adsorption properties of fly ash ascribed to its chemical composition, high porosity, surface area and chemical composition (Kamboh *et al.*, 2009). The major compositions of fly ash are oxides of silica, iron and alumina are good coagulants for the decrease in concentration of toxic contaminants in wastewater. The adsorption characteristics of fly ash resulted from the high carbon content with specific surface area ranged from 2,000 to 6,800 cm^2/g (Shah *et al.*, 2015; Sanas *et al.*, 2016).

Effect of flow rate on removal efficiency: With increasing the flow rate the decreasing in the removal of BOD, COD, TN, TP and TC were observed (Figure 4). The reduction rate of these pollutants increments with the increase in adsorbent contact time. Sanas *et al.* (2016) found the same finding when applying different rates for COD removal. The results indicated that either FF or CBD approach showed higher removal efficiency at 0.5l/min. At flow rate 0.5 l/min the removal efficiency of BDF was 78%, 90%, 59%, 97% and 91%, while for FF approach was 49%, 48%, 52%, 97% and 86% for BOD, COD, TN, TP and TC, respectively. BDF approach showed higher removal efficiency in comparison with the FF approach for the studied pollutants. Since the adsorption capacity of briquette fly ash as well as the coagulation step (removes of BOD, COD, TN, TP and TC by 25%, 27%, 5%, 60% and 80%, respectively). The motive of increasing removal efficiency while the flow rate decreased for the reason that the sorption of pollutants rely upon three steps which are the transport of the pollutant from the bulk solution to the sorbent surface; adsorption on the particle surface; and transport within the sorbent particle (Barakat, 2010). Increasing the adsorption constant time between wastewater and fly ash by decreasing the flow rate allow for the three steps of sorption to occur without recovery.

Table 3. Comparison between FF (filter powder fly ash) and CBD (Jar test followed briquette bed) on chemical and microbiological wastewater quality of Janag drain (average n=4).

	Raw Water	FF	CBD	Article 51 ^a		Raw Water	FF	CBD	Article 51 ^a
	Mean±SD					Mean±SD			
pH	6.79 ^c ±0.11	7.41 ^b ±0.12	7.83 ^a ±0.14	6.5-8.5	Chloride	111.2 ^a ±4.8	108.3 ^a ±4.6	107.3 ^a ±4.6	
% Removal		-9.06	-15.18		% Removal	2.59	3.50		
Bicarbonate	505.4 ^a ±31.94	325.4 ^b ±20	267.4 ^c ±17		Nitrate	<0.2	<0.2	<0.2	
% Removal		35.62	47.09		% Removal	0.00	0.00		
TDS	840.8 ^c ±4.3	865.2 ^a ±4.3	858.8 ^b ±4.3	1000	TP	15.8 ^a ±1.3	0.63 ^b ±0.1	0.32 ^b ±0.026	
% Removal		-2.90	-2.14		% Removal	96.00	98.00		
TSS	60.4 ^a ±3.5	36.8 ^b ±4.1	26 ^c ±1.6		Sulphate	62.3 ^a ±1.7	54.2 ^b ±1.7	63.3 ^a ±1.7	
% Removal		39.1	56.95		% Removal	13	-2.00		
BOD	90 ^a ±7.9	35.02 ^b ±3.1	19.6 ^c ±2.1	30	Cadmium	<0.2 ^a	<0.2 ^a	<0.2 ^a	0.03
% Removal		61.09	78.22		% Removal	0.00	0.00		
COD	269 ^a ±8.2	102 ^b ±2.8	26 ^c ±0.5	50	Chromium	0.011 ^a ±0.0015	0.002 ^b ±0.0001	<0.001 ^c	0.05
% Removal		61.93	90.48		% Removal	81.80	100.00		
Ammonia	41.5 ^a ±6.4	20.3 ^b ±4.4	18.5 ^c ±4.5		Copper	0.076 ^a ±0.0004	0.0209 ^b ±0.0018	0.076 ^a ±0.007	1
% Removal		51.1	55.4		% Removal	72.50	0.00		
TN	34.1 ^a ±10.4	16.4 ^b ±25.4	14.0 ^c ±4.4	15	Iron	0.78 ^a ±0.043	0.0805 ^c ±0.0044	0.652 ^b ±0.1628	5
% Removal		47.21	58.49		% Removal	89.70	16.00		
Calcium	77.5 ^b ±2.2	177.1 ^a ±4.9	78.0 ^b ±2.2		Lead	<0.2 ^a	<0.2 ^a	<0.2 ^a	0.01
% Removal		-128.69	-0.71		% Removal	0.00	0.00		
Potassium	28.7 ^a ±0.9	26.4 ^b ±0.9	28.7 ^a ±0.9		Manganese	1.396 ^a ±0.0015	1.263 ^b ±0.0013	1.002 ^c ±0.0012	2
% Removal		8.14	0.00		% Removal	9.53	28.22		
Magnesium	19.3 ^b ±0.4	21.1 ^a ±0.4	19.2 ^b ±0.4		Zinc	0.026 ^a ±0.03	0.011 ^c ±0.009	0.0153 ^b ±0.016	2
% Removal		-9.12	0.83		% Removal	57.71	41.20		
Sodium	151.6 ^a ±2	151.6 ^a ±2	152.6 ^a ±2		TC	700×10 ¹³ ±71×10 ⁴	100×10 ¹³ ±10×10 ⁴	66×10 ¹³ ±4×10 ⁴	5000
% Removal		0.00	-0.66		% Removal	85.7	90.5		

Negative values represent byproducts of treatment process or increase the concentration after application of treatment. pH is unitless, Total Coliform (TC) unit is CFU/100 ml, while the units of other parameters in table 3 are mg/L. Data represented as mean±SD of 4 sample. Means with single letter (A, B, C) in the same raw are significant (p<0.05). * article 51 of Law 48/1982

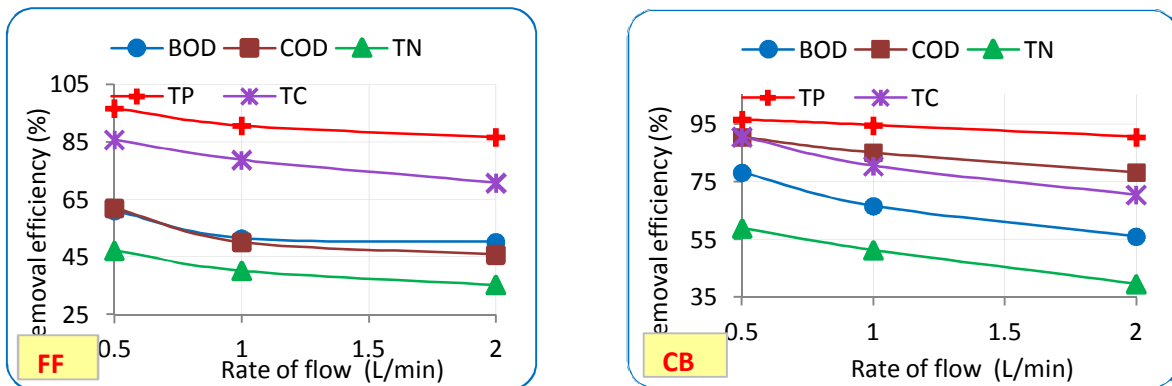


Figure 4. Effect of flow rate on BOD, COD, TN, TP and TC removal efficiency of FF (filter powder fly ash) and (CBD (briquette bed coupled with jar test).

Disinfection using chlorination:

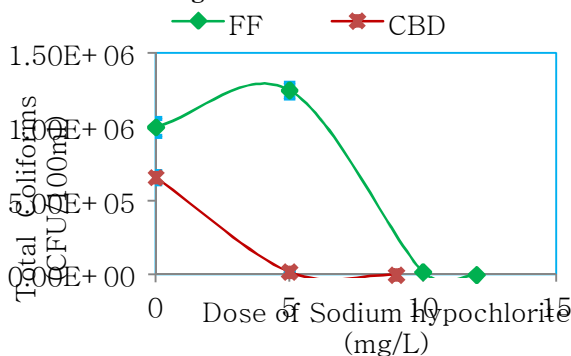


Figure 5. Effect of chlorine dose on total Coliforms count (CFU/100ml), where FF (filter powder fly ash) and CBD (briquette bed coupled with jar test).

The results of Total Coliforms (TC) and doses applied were previewed in figure 5. The results indicated as the dose increase the counts of TC decreases. The dose required to get rid of TC was 12 and 9 mg/l for water treated with FF and CBD, respectively.

4 Conclusion and Recommendation

The study of chemical characteristics of wastewater gives a considerable insight on drainage, industrial and municipal wastewater quality at Janag Drain, Kafr El-Zayat area depending on Law 48 for 1982 and its modification in 2013. The results indicated that the mixed drainage samples showed significantly different variation for all water quality parameters except pH with respect to drainage,

industrial and sewage wastewater. For Janag drain had BOD, COD, TN, TP & TC, while industrial wastewater had pH, TDS, TSS, BOD, COD & TC and municipal wastewater had TC higher the permissible limits.

The result of two approaches applied using fly ash indicated that the use of the briquette bed approach is more efficient for remove wastewater contaminant than fly ash filter powder (FF). The results of treatment by fly ash briquette bed proved that fly ash was capable of reducing BOD, COD, TN and TP by 77%, 90%, 59% and 100 %, respectively at flow rate 0.5 L/min to lower than the recommended limit. The chlorine doses required for disinfecting treated water from the CBD and FF approaches were 9 and 12 mg/L, respectively. Problems of environmental pollution can also be minimized by utilization of fly ash briquette bed approach in treatment of COD, BOD, TN, TP and total Coliforms (TC) from waste water. Further research needs attention on fly ash briquette bed approach followed by a disinfection process for utilization in a better way in situ.

Acknowledgement:

This study has been supported by the National Water Research Center (NWRC). The authors are also grateful to Director of Central Laboratory for Environmental Quality Monitoring (CLEQM) for the assisting and giving us a chance to complete this study. The authors are most appreciative to the working staff of CLEQM for their valuable cooperation.

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